Supplemental Material: Antibubbles dynamics: the drainage of an air film with incompressible interfaces

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"EXTENSIONAL SURFACE VISCOSITY": SURFACE/BULK ANALOGY FOR INCOMPRESSIBLE FLOWS

In our manuscript, we argue in the introduction that, provided the surfactant concentration is much above the cmc, and if the time for surfactant molecules to be adsorbed at the interface is much smaller than the time for these molecules to be advected along the interface, the surface density of surfactants remains constant, hence our hypothesis of an incompressible interface. In such a situation, elastic and viscous dilational effects are absent and the only component of viscosity that is relevant is the surface shear viscosity, exactly as it is the case in a tri-dimensional incompressible flow.

Now, surface velocity gradients along the flow direction are undoubtedly present due to the drainage and should be maximum near the poles of the antibubble. Nevertheless, these gradients should not have other effect for an incompressible interface than streamwise viscous dissipation, since there is no azimuthal velocity gradient. Streamwise viscous dissipation is important in many viscous flow situations, such as in the dynamics of solitary waves in falling liquid films [1], but the best analogy we can make with a three-dimensional flow would be the extensional flow describing the stretching of a viscous sheet with stress-free interfaces, like in the film casting or the float glass processes [2]. In an extensional flow, the viscous dissipation is associated to the velocity gradient in the flow direction, and the so-called extensional viscosity is 4 times larger than the bulk shear viscosity. The factor 4 is called the Trouton ratio and arises from the integration of the second-order parabolic correction of the uniform velocity profile across the liquid sheet. The analogy is straightforward for an extensional surface flow, except that the "extensional surface viscosity" is identical to the surface shear viscosity, i.e. no Trouton ratio, since the interface has no thickness in the frame of continuum mechanics.

MODEL DERIVATION

We develop here the lubrication model that describes the dynamics of an air film in antibubbles with incompressible interfaces.

Denoting $\hat{u}(\theta,t)=\frac{1}{h}\int_0^h u(r,\theta,t)\,\mathrm{d}r$ the average air velocity across the film, where u is the velocity field and $r=r'-R$ the coordinate inside the film (see Fig. 1), the mass conservation equation of the air flow is

$$\frac{\partial h}{\partial t}+\frac{1}{R\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\; h\hat{u}\right)=0\,. \tag{1}$$

Because the film thickness is of micron-scale [3], the smallness of the aspect ratio $\epsilon=h_0/(\pi R)\sim 10^{-4}$, where h_0 is the average initial thickness, enables the reduction of the momentum balance to the lubrication equation,

$$\mu_{\mathrm{air}}\frac{\partial^2 u}{\partial r^2}=\frac{1}{R}\frac{\partial p}{\partial\theta}\,, \tag{2}$$

where μ_{air} is the dynamic viscosity of air and $p(\theta,t)$ the pressure field, which remains uniform across the film. Since the slope of the interfaces remains very small everywhere, i.e. $(1/R)\partial_\theta h\sim\epsilon$, one also have that both surface velocities are identical at leading order, i.e. $u|_{r=0}=u|_{r=h}=u_{\mathrm{s}}(\theta,t)$. Integrating (2) three times thus yields

$$\hat{u}(\theta,t)=u_{\mathrm{s}}-\frac{h^2}{12\mu_{\mathrm{air}}R}\frac{\partial p}{\partial\theta}\,. \tag{3}$$

The flow being driven by the hydrostatic pressure difference between the poles, one can deduce from (3) the characteristic timescale for drainage, $\tau=\mu_{\mathrm{air}}\pi^2R/(\rho g h_0^2)\sim 100$ s, and so the scale for the air velocity, $u_0=\pi R/\tau\sim 100$ μm/s. The non-dimensionalization is performed using the following scaling: $r=h_0\bar{r}$, $h=h_0\bar{h}$, $t=\tau\bar{t}$, $\theta=\pi\bar{\theta}$, $u=u_0\bar{u}$, and $p=p_0\bar{p}$. The dimensionless conservation equation (1), combined with (3), then becomes

$$\frac{\partial\bar{h}}{\partial\bar{t}}+\frac{1}{\sin(\pi\bar{\theta})}\frac{\partial}{\partial\bar{\theta}}\left[\bar{h}\sin(\pi\bar{\theta})\left(\bar{u}_{\mathrm{s}}-\frac{\bar{h}^2}{6}\frac{\partial\bar{p}}{\partial\bar{\theta}}\right)\right]=0\,. \tag{4}$$

The dynamic surface stress condition at the outer air-liquid interface, neglecting inertia and mass transfer, is

$$\mathbf{n}\cdot\left[(p_{\mathrm{liq}}-p)\mathbf{I}+\mathbf{P}-\mathbf{P}_{\mathrm{liq}}\right]=\boldsymbol{\nabla}_{\mathrm{s}}\cdot(\gamma\mathbf{I}_{\mathrm{s}}+\mathbf{S}_{\mathrm{s}})\;, \tag{5}$$

where $\mathbf{n}$ is the normal vector as sketched in Fig. 1 of the paper, $\mathbf{P}$ is the viscous stress tensor, $\boldsymbol{\nabla}_{\mathrm{s}}=\mathbf{I}_{\mathrm{s}}\cdot\boldsymbol{\nabla}$ is the surface gradient operator, $\mathbf{I}_{\mathrm{s}}=(\mathbf{I}-\mathbf{nn})$ is the surface identity tensor and $\mathbf{S}_{\mathrm{s}}$ is the surface stress tensor. For sufficiently slow rates of deformation, the stress-deformation behavior of a fluid-fluid interface can be characterized by the linear Boussinesq-Scriven constitutive surface fluid model [4],

$$\mathbf{S}_{\mathrm{s}}=(\kappa-\varepsilon)\boldsymbol{\nabla}_{\mathrm{s}}\cdot\mathbf{u}_{\mathrm{s}}\,\mathbf{I}_{\mathrm{s}}+2\varepsilon\mathbf{D}_{\mathrm{s}}\;, \tag{6}$$

where κ and ε are, respectively, the surface dilational and shear viscosities, and $\mathbf{D}_\mathrm{s} = \left[(\boldsymbol{\nabla}_\mathrm{s}\mathbf{u}_\mathrm{s})\cdot\mathbf{I}_\mathrm{s} + \mathbf{I}_\mathrm{s}\cdot(\boldsymbol{\nabla}_\mathrm{s}\mathbf{u}_\mathrm{s})^T\right]$ is the surface rate of deformation tensor. Next, we emphasize the following central hypotheses:

H1: As a result of the lubrication approximation, the interfacial viscous shear stress exerted by the surrounding liquid can be neglected as compared to the one exerted by the gas, i.e. $\mathbf{P}_\mathrm{liq} \ll \mathbf{P}$. This can be explained by the fact that $\mu_\mathrm{liq}/\delta \ll \mu_\mathrm{air}/h_0$, where $\delta \sim \sqrt{\mu_\mathrm{liq}\tau/\rho}$ is the thickness of the viscous boundary layer in the liquid phase [5]; for $\mu_\mathrm{liq} \sim 10^{-3}$ Pa.s and $\rho \sim 1000$ kg/m^3, $\delta \sim 10^{-2}$ m, which is comparable to the antibubble radius R, as expected for a Stokes flow, hence using $\mu_\mathrm{air} = 1.85\times 10^{-5}$ Pa.s, we get $\epsilon\mu_\mathrm{liq}/\mu_\mathrm{air} \ll 1$.

H2: We assume the liquid concentration to be much above the cmc such that surfactants are always available in the sublayer of the continuous phase to replenish the interface. Additionally, we assume that the adsorption rate of surfactant is much higher than the interfacial strain rate, or equivalently $\tau_\mathrm{ads} \ll \tau_\mathrm{adv} \sim \tau$. Under these assumptions, the surface density remains constant, which excludes surface elasticity effects, i.e. $\boldsymbol{\nabla}_\mathrm{s}\gamma = 0$, and confers to the interface an incompressible nature, i.e. $\boldsymbol{\nabla}_\mathrm{s}\cdot\mathbf{u}_\mathrm{s} = 0$. Consequently, only the surface shear viscosity ε remains in the model.

H3: We further assume a "Newtonian" interface, i.e. a constant surface shear viscosity, which is reasonable for low surface shear rates, i.e. $1/\tau \sim 0.01$Hz (see Fig. 6).

Under the above assumptions and given that $\boldsymbol{\nabla}_\mathrm{s}\cdot\mathbf{I}_\mathrm{s} = 2H\mathbf{n}$ where H is the mean curvature, (5) becomes

$$\mathbf{n}\cdot[(p_\mathrm{liq}-p)\mathbf{I}+\mathbf{P}] = 2H\gamma\mathbf{n} + 2\varepsilon\boldsymbol{\nabla}_\mathrm{s}\cdot\mathbf{D}_\mathrm{s}\,. \tag{7}$$

The mean curvature not only depends on the sphericity of the antibubble, but also on local variations of the film curvature, such that, in the frame of the lubrication approximation, $H = \frac{1}{R} - \frac{1}{2}\boldsymbol{\nabla}_\mathrm{s}{}^2 h$. Next, the liquid pressure depends on the reference pressure p_ref taken at the North pole (see Fig. 1) as well as on the hydrostatic pressure along the air film, such that $p_\mathrm{liq} - p_\mathrm{ref} = \rho g(2R-z) = \rho g R\,(1+\cos\theta)$. Taking the normal projection of (7) at leading-order of the lubrication approximation, the pressure in the air film is

$$p = p_\mathrm{ref} + \rho g R(1+\cos\theta) + \gamma\left(\frac{2}{R} - \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left[\sin\theta\frac{\partial h}{\partial\theta}\right]\right). \tag{8}$$

Here it remains to account for the molecular interactions responsible for the ultimate rupture of antibubbles. In the frame of the lubrication theory, it can be expressed as a correction to the pressure field via the transformation $p \to p - \phi(h)$ where $\phi = A'/(6\pi h^3)$ is the disjoining pressure, and A' is the Hamaker constant [6]. The dimensionless pressure gradient finally takes the form

$$\frac{\partial\bar{p}}{\partial\bar{\theta}} = \frac{1}{2}\frac{\partial}{\partial\bar{\theta}}\left[\cos(\pi\bar{\theta}) - \frac{Bo}{\sin(\pi\bar{\theta})}\frac{\partial}{\partial\bar{\theta}}\left(\sin(\pi\bar{\theta})\frac{\partial\bar{h}}{\partial\bar{\theta}}\right) + \frac{A}{\bar{h}^3}\right], \tag{9}$$

where $Bo = \gamma h_0/(\rho g\pi^2 R^3)$ is the Bond number and $A = A'/6\pi\rho g R h_0^3$ is the dimensionless Hamaker constant.

The tangential projection of (7) at leading order becomes in dimensional and dimensionless forms (see [4]), respectively,

$$\mu_\mathrm{air}\left.\frac{\partial u}{\partial r}\right|_{\frac{h}{2}} = 2\frac{\varepsilon}{R^2}\left[\frac{\partial}{\partial\theta}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}(\sin\theta\, u_\mathrm{s})\right) + u_\mathrm{s}\right], \tag{10}$$

$$\bar{h}\frac{\partial\bar{p}}{\partial\bar{\theta}} = 2Bq\left[\frac{\partial}{\partial\bar{\theta}}\left(\frac{1}{\sin(\pi\bar{\theta})}\frac{\partial}{\partial\bar{\theta}}(\sin(\pi\bar{\theta})\,\bar{u}_\mathrm{s})\right) + \pi^2\bar{u}_\mathrm{s}\right], \tag{11}$$

where $Bq = \varepsilon h_0/(\mu_\mathrm{air}\pi^2 R^2)$ is the Boussinesq number.

LIFETIME IN THE NO-SLIP LIMIT

Since from simulations, we observed that the film thins primarily at the South pole where $\bar{h} = \bar{h}_\mathrm{min}$, we can rewrite (4) in the limit of $\bar{\theta} \to 0$, in the no-slip limit $u_\mathrm{s} = 0$ (or equivalently $Bq \to \infty$), considering no Van der Waals effect $A = 0$ and with neglecting curvature gradient effects $Bo = 0$. The result is an ordinary differential equation (ODE), which is space-independent:

$$\frac{\mathrm{d}\bar{h}_\mathrm{min}}{\mathrm{d}\bar{t}} + \frac{\pi^2\bar{h}_\mathrm{min}^3}{6} = 0\,, \tag{12}$$

whose solution is

$$\bar{h}_\mathrm{min}(\bar{t}) = \left(1 + \frac{\pi^2}{3}\,\bar{t}\right)^{-1/2}. \tag{13}$$

The lifetime at rupture, i.e. for $h_\mathrm{min} = h_c$, thus becomes

$$t_\mathrm{life} = \frac{3\mu_\mathrm{air}R\left(1-\frac{h_c^2}{h_0^2}\right)}{\rho g h_c^2}\,, \tag{14}$$

as (5) in the paper.

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